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Low-temperature antiferromagnetic ordering in the heavy-fermion metal YbPd

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Abstract. We investigate the low-temperature properties of YbPd by specific heat and thermal expansion measurements down to 0.1 K. This material crystallizes in a cubic CsCl-structure and four phase transitions at 125, 105, 1.9 and 0.5 K have been reported previously. The two transitions at higher temperature were suspected to be of structural origin, whereas the two low-T transitions are magnetic, confirmed by susceptibility measurements and Mössbauer spectroscopy. Our low temperature specific heat and thermal expansion data prove antiferromagnetic ordering at T_N=1.9 K and an additional first-order antiferromagnetic transition at 0.6 K. The entropy reaches approximately half of Rln2 at 5 K, confirming a doublet ground state. The enhanced value of the Sommerfeld coefficient at 0.1 K suggests a classification of YbPd as a magnetically ordered heavy-fermion metal.

The cubic system YbPd, crystallizing in the CsCl-structure, has provided puzzling experimental observations. It exhibits four transitions below room temperature at 125, 105, 1.9 and 0.5 K [1]. At the higher two transition temperatures, specific heat and thermal expansion show two symmetric peaks and two sharp spikes, respectively, implying first-order structural phase transitions, although a possible tetragonal distortion at 4.2 K is very small [2]. Furthermore, the ^{170}Yb Mössbauer measurements [2] detected no asymmetric spectrum at 4.2 K, showing Yb nuclei experiencing no electric field gradient, and thus, behaving essentially cubic-like. The most puzzling behavior is found in the phase below the transition temperature of 1.9 K. AC-susceptibility shows a kink and ^{170}Yb Mössbauer measurements confirm its magnetic origin. The Mössbauer study demonstrates that only one half of the Yb-atoms experiences the hyperfine field due to the magnetic order while the other half stays non-magnetic. We note, however, that a uniform tetragonal distortion would not produce crystallographically inequivalent Yb sites. The study concluded that the system possesses two Yb charge states with equal fraction, one magnetic, causing the magnetic transition and the other one non-magnetic Yb^{2+}. This is reminiscent of the charge order with Yb^{2+} and Yb^{3+} sites in Yb_4As_3, accompanied by a trigonal distortion along [111] [3]. Here, a trigonal distortion produces two crystallographically inequivalent Yb sites, resulting in an unequal fraction of 1:3 for Yb^{3+}:Yb^{2+}. Finally, the transition at 0.6 K in YbPd is less studied. AC-susceptibility exhibits an anomaly with a hysteresis, indicating a first order magnetic transition. In this paper, we examine the scenario of two different Yb charge states by measuring specific heat at low temperatures and obtaining its magnetic entropy. We also focus on thermal expansion to get further insight on the two phase transitions at low temperatures.

Polycrystalline samples were prepared by melting the high-purity elements with 3% excess of Yb in closed Ta-crucibles. The formation of a single phase was confirmed by X-ray

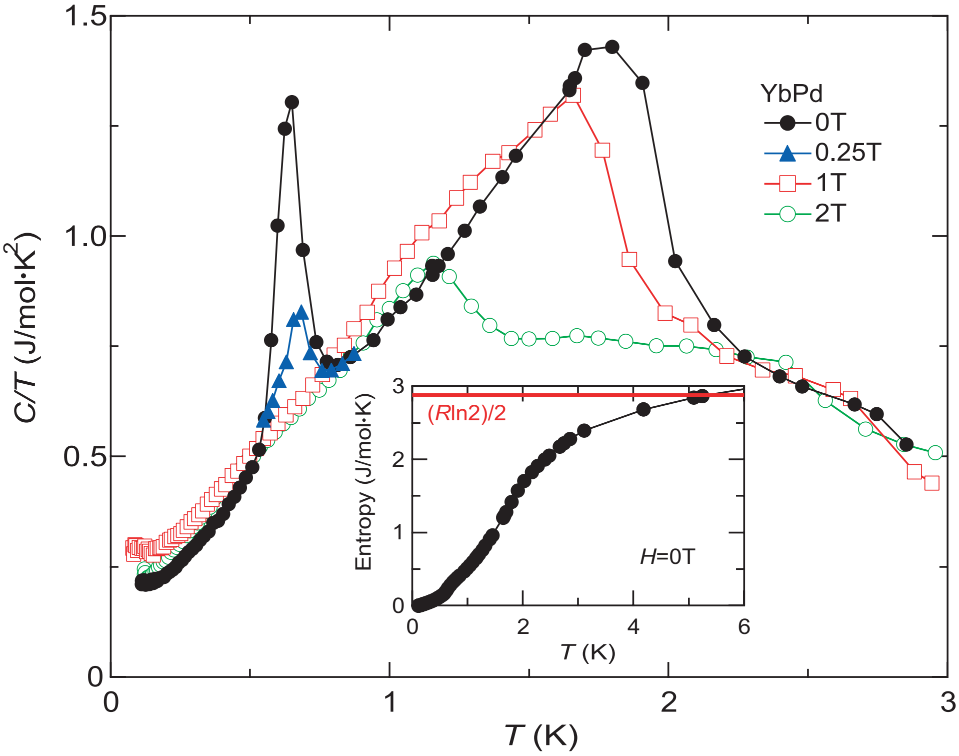

Figure 1. Specific heat divided by temperature, C/T, of YbPd as a function of temperature at different magnetic fields. Inset: magnetic entropy at zero-field.

diffraction measurements. Electrical resistivity and specific heat measurements above 4.2 K are in close agreement to Ref. [1] and reproduce the phase transitions at 130 and 110 K. The low-temperature specific heat and the thermal expansion have been determined in a dilution refrigerator by utilizing the quasiadiabatic heat pulse technique and an ultrahigh resolution capacitive dilatometer, respectively.

Figure 1 shows the specific heat of YbPd at low temperatures in different fields. The zero-field data exhibit two anomalies at 1.9 and 0.6 K, in agreement with the reported studies. The one at 1.9 K has a lambda-like shape, while the other one at 0.6 K shows a sharp and more symmetric anomaly, indicative of a first order transition. Magnetic field shifts the transition temperature of 1.9 K to lower temperatures, indicating antiferromagnetic order. The anomaly at 0.6 K is rapidly suppressed by magnetic field, however, the transition temperature stays constant. Notably, the residual C/T as $T \to 0\,K$ is enhanced, around 0.22 J/mol·K^2, which classifies YbPd as heavy fermion system. From the zero-field $C(T)/T$ data, we determined the magnetic entropy shown in the inset. Above the transition at 1.9 K it tends to saturate and reaches half of Rln2 at 5 K. This result can be understood by assuming one half of the Yb sites being in a nonmagnetic configuration. The other half contributes as heavy fermion antiferromagnet with a doublet ground state and Kondo interaction reducing the entropy below $0.5R\ln 2$ at 1.9 K.

Next, we discuss the linear thermal expansion, $\Delta L/L$, of our YbPd polycrystals. $\Delta L/L$ is negative in the entire measured temperature range up to 4 K, indicating the sample shrinking with increasing temperature. Such behavior is expected for Yb-based heavy-fermion systems and results from the negative pressure dependence of the Kondo temperature. At 1.9 K, it changes its slope, corresponding to a smeared step in thermal expansion coefficient $\alpha(T)$. $\Delta L/L$ shows a step for the transition at 0.6 K. The hysteresis between heating and cooling is due to the first

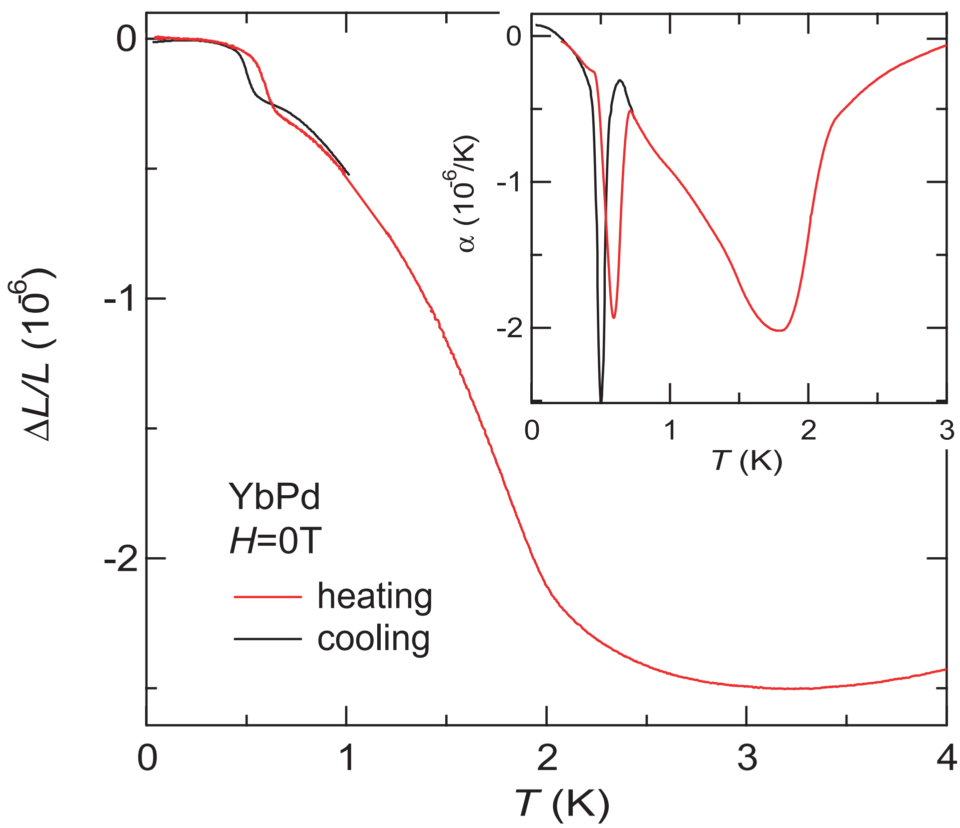

Figure 2. Thermal expansion of YbPd at zero-field. Red and black curves are for heating and cooling the sample. Inset: thermal expansion coefficient of YbPd.

order nature of the transition.

In conclusion, we have studied the low temperature phases with transitions at 1.9 and 0.6 K in YbPd by measuring specific heat and thermal expansion. The obtained magnetic entropy at 1.9 K is smaller than one half of $R\ln 2$. This confirms a doublet crystal electric field ground state, excluding the possibility of a Γ_8 quartet, proposed in Ref. [4], and also supports the interpretation of the Mössbauer spectroscopy experiments that only half of the Yb sites is in a magnetic configuration [2]. We observed a large residual Sommerfeld coefficient as $T \to 0$, indicating heavy fermion behavior. Within the picture of a 1:1 fraction of magnetic and non-magnetic Yb ions, our results suggest that the magnetic ones form a heavy fermion antiferromagnet. The magnetic transitions have also been confirmed by thermal expansion.

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